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TITLE : ACETAL RESIN COMPOSITION

ABSTRACT : PURPOSE: To provide an acetal resin composition capable of giving a molded product excellent in surface appearance, obtained by adding at least one specific compound of alkali (alkaline earth) metal and at least one of organotitanate and organosilane to an acetal resin.

CONSTITUTION: At least one of hydroxide, inorg. acid salt, carboxylate and alkoxide of Na, K, Mg, Ca and Ba (A) and at least one of organotitanate (e.g. isopropyltriacyrltitanate) and organosilane (e.g.  $\gamma$ -chloropropyltrimethoxysilane) (B) are added to an acetal resin in a weight ratio of about 100:0.1-20. That is, compounds A and B are compounded in 100pts.wt. acetal resin in a ratio of about 0.004-5pts.wt. as a total amount and the obtained compound is kneaded by using an extruder or a kneader to obtain an acetal resin composition.

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## DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention improves the compatibility between the resin of the polymer blend which consists of thermoplastic polyacetal resin and polyolefine system resin, and it relates to the resin constituent which realized shock-proof amelioration, without spoiling the mechanical and thermal property of polyacetal resin.

[0002]

[Description of the Prior Art] Polyacetal resin has the property which was excellent in a mechanical and thermal property, electrical characteristics, sliding nature, a moldability, the dimensional stability of mold goods, etc., and is widely used for an electrical machinery and apparatus, autoparts, a precision machinery component, etc. as a structural material, a mechanism element, etc. however -- for example, when it uses for the application for which high shock resistance is needed like housing of home electronics, field impact strength is inadequate -- etc. -- it had the fault. In order to improve such a fault, the approach of blending rubber-like components, such as polyurethane and an olefin system elastomer, conventionally has been tried. However, since the compatibility of the ERASUMA component and polyacetal resin which were added is not good, and it is inadequate just to have carried out melting kneading simply as for the interface bond strength between both resin, a maldistribution occurs, the property which was excellent in polyacetal resin is spoiled, surface exfoliation of mold goods is caused, or there are problems -- shock resistance is not obtained enough. Although amelioration of blending with polyacetal resin the copolymer which introduced the comonomer component with polar methyl methacrylate etc. into polyolefin resin in order to conquer this problem, and the resin which contains a reactant glycidyl group in the molecular structure was performed, it will not have resulted, by the time it is put in practical use by the shock-proof improvement of polyacetal being inadequate.

[0003]

[Means for Solving the Problem] Then, this invention person etc. is blending the catalyst for promoting these reactions with the polyolefine system resin which has the acid-anhydride radical which is rich in reactivity in thermoplastic polyacetal resin as a result of examining this point wholeheartedly, and improved the compatibility of polyacetal resin and polyolefine system resin, and without spoiling the mechanical and thermal property which was excellent in polyacetal resin, it succeeds in considering as the shaping resin ingredient to which shock resistance was added, and came to complete this invention. That is, this invention is (A). Thermoplastic polyacetal resin 100 It is (B) to the weight section. Acid-anhydride denaturation polyolefine system resin 1-100 The weight section and (C) The esterification reaction catalyst 0.01 - 5 weight sections, and (D) One or more sorts of bulking agents 0-150 chosen from a fibrous bulking agent and powder-like bulking agent, a tabular bulking agent, and a hollow-like bulking agent It is the polyacetal resin constituent which blends the weight section and changes.

[0004] Hereafter, the constituent of this invention is explained. The polyacetal resin in this invention is an oxy-methylene unit (-CH<sub>2</sub>O-). It is thermoplastics which uses as a principal component the main high molecular compound repeatedly made into a configuration unit. What homopolymerized

formaldehyde or a trioxane, tetra-oxane, etc. by the usual approach, Or the copolymer which consists of these two or more sorts or this monomer, and ethylene oxide, A copolymer with cyclic ester, such as cyclic ether, such as propylene oxide, OKISA cyclobutane, 1, and 3-dioxolane, beta propiolactone, and gamma-butyrolactone, or a vinyl compound of a certain kind is (A). It is used as a component. Moreover, as for this polyacetal resin, a part of the molecule end may be changed into ether linkage, an ester bond, etc. Furthermore, if a part or all of this polyacetal resin is polyacetal resin with many hydroxyl-group contents manufactured by the approach as shown below (wealth hydroxyl-group polyacetal resin is called hereafter), it is still more desirable and, in this case, the effectiveness of this invention is more remarkable. Moreover, the hydroxyl-group content in polyacetal resin is 30 mmol/kg in this case. If it becomes above, a desirable result will be obtained especially. as the approach of manufacturing the above-mentioned wealth hydroxyl-group polyacetal resin -- a trioxane -- BF3 etc. -- although there is the approach of carrying out little addition of the compound containing hydroxyl groups, such as water, ethylene glycol, a glycerol, and glycidol, etc. in case a polymerization is performed in a cation initiator, especially the process is not limited. Moreover, copolymerization types, such as the degree of polymerization or existence of branching, a homopolymer, a copolymer or random, a block, and a graft, are not asked. Moreover, especially the binding site of a hydroxyl group is not restricted, either.

[0005] (B) of this invention A component is acid-anhydride denaturation polyolefine system resin which denaturalized polyolefine system resin with the acid anhydride. As polyolefine system resin used here Ethylene, a propylene, a butene, a hexene, octene, nonene, decene, The homopolymer of alpha olefins, such as dodecen, or the random which consists of these two or more sorts, To a block, a graft copolymer, or these, 1, 4-hexadiene, Nonconjugated diene, such as dicyclopentadiene, 5-ethylidene-2-norbornene, 2, and 5-NORUBONA diene, Conjugated diene components, such as a butadiene, an isoprene, and piperylene, an acrylic acid, Derivatives, such as alpha, such as a methacrylic acid, beta partial saturation acid, or its ester, acrylonitrile, Vinyl ester, such as aromatic series vinyl compounds, such as styrene and alpha methyl styrene, or vinyl acetate, The random which changes including one or more sorts in comonomer components, such as vinyl ether, such as vinyl methyl ether, and a derivative of these vinyl system compounds, a block, or a graft copolymer is mentioned, and how, such as the polymerization degree, a side chain, the existence of branching or extent, and a copolymerization presentation ratio, is not asked. Moreover, as an acid anhydride used for denaturation, one or more sorts of things chosen from unsaturated-carboxylic-acid anhydrides, such as a maleic anhydride, an anhydrous citraconic acid, itaconic acid anhydride, tetrahydro phthalic anhydride, an anhydrous NAJIKKU acid, an anhydrous methyl NAJIKKU acid, and an allyl compound succinic anhydride, or the derivative of those are used. Moreover, although the approach to which heat unsaturated-carboxylic-acid anhydrides, such as polyolefine system resin and a maleic anhydride, or the derivative of those with radical initiators, such as suitable organic peroxide, in the state of a solution condition or melting, and it is made to react as the denaturation approach is suitable, especially the manufacturing method is not limited. Here, the loadings of both components are polyolefine system resin 100. An acid anhydride 0.1 - 30 weight sections are suitable to the weight section. Since the compatibility between polyacetal resin and polyolefine system resin is not fully improved when there are too few amounts of the effective acid anhydride in acid-anhydride denaturation polyolefine system resin, the effectiveness of this invention is not acquired, but when many [ too ], it may become the cause of a lifting, poor dispersibility, and poor shaping about gelation. moreover, (B) the loadings of a component -- (A) Component 100 the weight section -- receiving -- 1-100 the weight section -- it is 5 - 50 weight section preferably. (B) When the loadings of a component are too low, it is not fully demonstrated, but the effectiveness of this invention will spoil the property of polyacetal resin, when many [ too much ].

[0006] (C) used by this invention As a catalyst for esterification reaction promotion which is a component Pyridine derivatives and those salts, such as a pyridine, 4-dimethylaminopyridine, and 4-pyrrolidinopyridine, A triethylamine, trimethylamine, triethylenediamine, N, and N'-dimethyl piperidine, The third class amines and its derivatives, such as benzyl monomethylamine and dimethylaniline, Quarternary ammonium salt and its derivatives, such as trimethyl benzyl

ammoniumchloride, Aliphatic series organic acids, such as an acetic acid and a benzoic acid, an aromatic series organic acid, and sodium acetate, The salt of organic acids, such as sodium lauryl sulfate and Para toluenesulfonic acid sodium, Tetrabutyl zirconate, zirconium NAFUTETO, tetrabutyl titanate, Tetra-octyl titanate, tetra-phenyl tin, zinc acetate, the first tin of oxalic acid, Zinc naphthenate, iron acetyl acetate, manganese naphthenate; triphenyl antimony, Various kinds of metallic compounds of organometallic compounds, such as tributyl antimony, a triphenyl bismuth, and dibutyltin dichloride, a zinc chloride, magnesium titanate, and magnesium zirconate etc. are mentioned, and one or more of sorts of these can be added. As a desirable catalyst, pyridines, its derivative or those salts and tertiary amine, or its derivative is especially mentioned from fields, such as thermal stability and a hue. (C) The loadings of a component are (A). 0.01 - 5 weight section is suitable to the component 100 weight section. When there are too few amounts of catalysts, a reaction cannot fully progress and effectiveness of this invention cannot be acquired.

[0007] (D) used by this invention Although the bulking agent of a component is not necessarily an indispensable component, blending is desirable in order to obtain mold goods excellent in engine performance, such as a mechanical strength, thermal resistance, dimensional stability, and an electrical property. (D) As a component, the bulking agent of the shape of the shape of fibrous and a powder, tabular, or hollow is used according to the purpose. As a fibrous bulking agent, inorganic fiber-like matter, such as metal fiber-like objects, such as a glass fiber, an asbestos fiber, carbon fiber, silicon fiber, a silica alumina fiber, a zirconia fiber, a boron nitride fiber, nitriding silicon fiber, a boron fiber, titanate-acid potash fiber and also stainless steel, aluminum, titanium, copper, and brass, is mentioned, and especially a glass fiber or carbon fiber is used abundantly. In addition, high-melting organic fibrous material, such as aromatic polyamide resin, a fluororesin, and acrylic resin, can also be used. As a powder-like bulking agent, carbonates, such as carbon black, a silica, quartz powder, a glass bead, glass powder, calcium silicate, aluminum silicate, a kaolin, talc, clay, diatomite, the silic acid salt like a WORASU night, ferrous oxide, titanium oxide, the metallic oxide like an alumina, a calcium sulfate, a sulfate of the metal like a barium sulfate, a calcium carbonate, a magnesium carbonate, and a dolomite, other carbonization silicon, nitriding silicon, boron nitride, various metal powder, etc. are mentioned. As a tabular bulking agent, a mica, a glass flake, various metallic foils, etc. are mentioned, and milt balun, metal balun, glass balun, etc. are mentioned as a hollow-like bulking agent. Moreover, as these bulking agents, that by which the front face was processed by the organic silane, organic borane, organic titanate, urethane, etc. can also be used preferably. These bulking agents can use together and use one sort or two sorts or more. Especially concomitant use of a fibrous bulking agent especially a glass fiber or carbon fiber, and the shape of a powder and a tabular bulking agent is a combination desirable when it has a mechanical strength, dimensional accuracy, an electrical property, etc. (D) The loadings of a component are (A). Component 100 It is 150 to the weight section. If it is below the weight section and an addition becomes excessive, fabrication nature and toughness may be injured and it may not be desirable.

[0008] The constituent of this invention is adjusted by carrying out melting mixing of each component mentioned above by various approaches. For example, although melting kneading of the specified quantity of acetal resin, acid-anhydride denaturation polyolefine system resin, and a catalyst is carried out, and the approach of cutting to a pellet type is mentioned after cooling, about the combination stage of each component, or especially its approach, there is nothing what is limited. in addition, this time -- the need -- responding -- the increase of an antioxidant, an ultraviolet ray absorbent, an optical protective agent, a phosphite stabilizer, a peroxide decomposition agent, and a basic adjuvant -- a nucleating additive -- the amount of arbitration may be blended in the range which does not spoil the physical properties of this invention for additives, such as a plasticizer, lubricant, an antistatic agent, a flame retarder, and a coloring agent. Furthermore, it is possible to restrict to the range which does not spoil the physical properties to the constituent of this invention, and to blend other polymers suitably. In addition, the constituent obtained by this invention can be fabricated by the usual approach.

[0009]

[Effect of the Invention] Holding the property which was excellent in polyacetal resin in the thermal

property or the mechanical strength by improving the compatibility of polyacetal resin and polyolefine system resin, it improves shock resistance remarkably, and does not have the poor appearance of mold goods and surface exfoliation which are based badly [ distribution of distributed resin ], either, and the constituent obtained by this invention can expect the application to many applications.

[0010]

[Example] Although an example explains this invention still more concretely below, this invention is not limited to these.

Example 1 (A) Polyacetal resin 100 The weight section and polypropylene resin 100 (yes the Mitsui Petrochemical Industries, Ltd. make, pole J440) (B) which denaturalized the weight section in the maleic-anhydride 10 weight section They are 5 weight sections and (C) about maleic-anhydride denaturation polypropylene I. It is the dimethylamino pyridine 0.2 as a catalyst of a component. The weight section is mixed and they are laying temperature 190 \*\* and screw-speed 80rpm with a 30mm biaxial extruder. Melting kneading was carried out and it pelletized. Subsequently, from this pellet, it fabricated with the injection molding machine to the test piece, and evaluation of the following Izod impactive strength and a surface friction test was performed. An evaluation result is shown in Table 1. [Izod impactive strength] ASTM D256 It followed and impact strength (kgf-cm/cm) with a notch was measured.

[Surface friction test] After sticking the cellophane tape and tearing off on a test piece front face, viewing estimated the exfoliation situation and what was seen by O in what exfoliation was not regarded as was made into x.

It is (B) as shown in two to example 4 table 1. Except having changed the loadings of maleic-anhydride denaturation polypropylene I, the test piece was produced like the example 1 and it evaluated similarly. A presentation and an evaluation result are shown in Table 1.

the example of a comparison -- 1 polyacetal resin, the test piece was produced like the example 1 and independent combination estimated similarly. A presentation and an evaluation result are shown in Table 1.

Examples 2-4 of a comparison (B) A component and (C) Except having changed the loadings of a component, the test piece was produced like the example 1 and it evaluated similarly. A presentation and an evaluation result are shown in Table 1.

[0011] Example 5 (B) It replaces with maleic-anhydride denaturation polypropylene I, and is polypropylene 100 about the rate of maleic-anhydride denaturation. Except having carried out 20 weight sections combination of the maleic-anhydride denaturation polypropylene II made into 5 weight sections to the weight section, the test piece was produced like the example 2 and it evaluated similarly. A presentation and an evaluation result are shown in Table 2.

Example 6 (B) It replaces with maleic-anhydride denaturation polypropylene II, and is polypropylene 100 about the rate of maleic-anhydride denaturation. Maleic-anhydride denaturation polypropylene III made into 25 weight sections to the weight section Except having carried out 20 weight sections combination, the test piece was produced like the example 5 and it evaluated similarly. A presentation and an evaluation result are shown in Table 2.

Example 5 of a comparison (B) Maleic-anhydride denaturation polypropylene III It replaces with and is polypropylene 100. Except having carried out 20 weight sections combination of the maleic-acid denaturation polypropylene which denaturalized in the maleic-acid 10 weight section to the weight section, the test piece was produced like the example 6 and it evaluated similarly. A presentation and an evaluation result are shown in Table 2.

Example 6 of a comparison (B) Maleic-anhydride denaturation polypropylene III It replaced with, and except having blended the non-denaturalized polypropylene resin 20 weight section, the test piece was produced like the example 6 and it evaluated similarly. A presentation and an evaluation result are shown in Table 2.

[0012] Examples 7-8 (C) Except having changed the loadings of the dimethylamino pyridine which is a component, the test piece was produced like the example 2 and it evaluated similarly. A presentation and an evaluation result are shown in Table 3.

Examples 9-13 (C) Except having used triethylenediamine, trimethyl benzyl ammonium KORORAIDO, sodium acetate, tetrabutyl zirconate, or a zinc chloride as a component, the test piece was produced like the example 2 and it evaluated similarly. A presentation and an evaluation result are shown in Table 3.

[0013] Example 14 (B) It replaces with maleic-anhydride denaturation polypropylene I, and is ethylene propylene rubber (EPR) 100. Maleic-anhydride denaturation EPR 20 which denaturalized in the maleic-anhydride 10 weight section to the weight section Except having blended the weight section, the test piece was produced like the example 2 and it evaluated similarly. A presentation and an evaluation result are shown in Table 4.

Example 7 of a comparison (B) It replaced with the maleic-anhydride denaturation EPR, and except having used Native EPR, the test piece was produced like the example 14 and it evaluated similarly. A presentation and an evaluation result are shown in Table 4.

[0014] examples 15-17 and the examples 8-11 of a comparison -- further -- (D) Except having blended glass fiber as a component, the test piece was produced like said example and it evaluated similarly. A presentation and an evaluation result are shown in Table 5.

[0015] Examples 18-19 and examples 12-14 of a comparison (A) As a component, except having used the wealth hydroxyl-group polyacetal resin of hydroxyl-group content 100 mmol/kg, the test piece was produced like said example and it evaluated similarly. A presentation and an evaluation result are shown in Table 6.

examples 20-21 -- further -- (D) About the case where glass fiber is blended as a component, the test piece was produced like examples 18-19, and it evaluated similarly. A presentation and an evaluation result are shown in Table 6.

the examples 15-18 of a comparison -- further -- (D) About the case where glass fiber is blended as a component, the test piece was produced like the examples 12-14 of a comparison, and it evaluated similarly. A presentation and an evaluation result are shown in Table 6.

[0016] Examples 22-25 (D) When bulking agents other than glass fiber were blended as a component, about the case where glass fiber and a mica are used together, the test piece was produced like said example and it evaluated similarly. A presentation and an evaluation result are shown in Table 7.

Examples 19-22 of a comparison (B) It replaced with maleic-anhydride denaturation polypropylene I, and except having used native polypropylene, the test piece was produced like examples 22-25, and it evaluated similarly. A presentation and an evaluation result are shown in Table 7.

[0017]

[Table 1]

	単位	実施例 1	実施例 2	実施例 3	実施例 4	比較例 1	比較例 2	比較例 3	比較例 4
(A) ポリアセタール樹脂	重量部	100	100	100	100	100	100	100	100
(B) 無水マレイン酸変性ポリプロピレン I	重量部	5	20	40	80			120	20
(C) ジメチルアミノピリジン	重量部	0.2	0.2	0.2	0.2		0.2	0.4	
Izod衝撃強度	kgf cm/cm	11.5	12.1	14.8	18.2	6.5	6.4	13.1	5.7
表層剥離試験	—	○	○	○	○	○	○	×	×

[0018]

[Table 2]

	単位	実施例5	実施例6	比較例5	比較例6
(A) ポリアセタール樹脂	重量部	100	100	100	100
(B) 無水マレイン酸変性ポリプロピレンII	重量部	20			
(B) 無水マレイン酸変性ポリプロピレンIII	重量部		20		
(B') マレイン酸変性ポリプロピレン	重量部			20	
(B') 未変性ポリプロピレン	重量部				20
(C) ジメチルアミノピリジン	重量部	0.2	0.2	0.2	0.2
Izod衝撃強度	kgfcm/cm	11.6	13.6	5.8	3.5
表面剥離試験	—	○	○	×	×

[0019]

[Table 3]



	単位	実施例7	実施例8	実施例9	実施例10	実施例11	実施例12	実施例13
(A) ポリアセタール樹脂	重量部	100	100	100	100	100	100	100
(B) 無水マレイン酸変性ポリプロピレンI	重量部	20	20	20	20	20	20	20
(C) ジメチルアミノピリジン	重量部	0.03	4					
(C) トリエチレンジアミン	重量部			0.2				
(C) トリメチルベンジルアンモニウムクロライド	重量部				0.2			
(C) 酢酸ナトリウム	重量部					0.1		
(C) テトラブチルジルコネート	重量部						0.2	
(C) 塩化亜鉛	重量部							0.2
Izod衝撃強度	kgfcm/cm	11.7	13.3	12.0	10.3	9.8	10.7	10.1
表面状態試験	—	○	○	○	○	○	○	○

[0020]

[Table 4]

	単位	実施例14	比較例7
(A) ポリアセタール樹脂	重量部	100	100
(B) 無水マレイン酸変性EPR	重量部	20	
(B') 未変性EPR	重量部		20
(C) ジメチルアミノピリジン	重量部	0.2	0.2
Izod衝撃強度	kgfcm/cm	18.9	5.7
表面剝離試験	—	○	×

[0021]

[Table 5]

	単位	実施例15	実施例16	実施例17	比較例8	比較例9	比較例10	比較例11
(A) ポリアセタール樹脂	重量部	100	100	100	100	100	100	100
(B) 無水マレイン酸変性ポリプロピレンI	重量部	20	30	50				
(B') 未変性ポリプロピレン	重量部						20	50
(C) ジメチルアミノピリジン	重量部	0.2	0.2	0.3			0.2	0.2
(D) ガラスファイバー	重量部	20	40	80	20	80	20	80
Izod衝撃強度	kgfcm/cm	22.1	25.7	28.9	8.4	10.1	5.0	6.0
表面状態試験	—	○	○	○	○	×	×	×

[0022]

[Table 6]

	単位	実施例18	実施例19	比較例12	比較例13	比較例14	実施例20	実施例21	比較例15	比較例16	比較例17	比較例18
(A) 富水樹脂ポリアセタール樹脂	重量部	100	100	100	100	100	100	100	100	100	100	100
(B) 無水マレイン酸変性ポリプロピレンI	重量部	20	50				20	50				
(B') 未変性ポリプロピレン	重量部				20	50					20	50
(C) ジメチルアミノピリジン	重量部	0.2	0.3		0.2	0.3	0.2	0.3			0.2	0.3
(D) ガラスファイバー	重量部						30	60	30	80	30	60
Izod衝撃強度	kgfcm/cm	15.0	18.2	6.0	4.2	4.5	27.4	33.2	7.6	8.2	4.3	6.7
表面硬度試験	—	○	○	○	×	×	○	○	○	×	×	×

[0023]  
[Table 7]

	単位	実施例22	比較例19	実施例23	比較例20	実施例24	比較例21	実施例25	比較例22
(A) ポリアセタール樹脂	重量部	100	100	100	100	100	100	100	100
(B) 無水マレイン酸変性ポリプロピレン I	重量部	30		30		30		30	
(B') 未変性ポリプロピレン	重量部		30		30		30		30
(C) ジメチルアミノピリジン	重量部	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
(D) ガラスファイバー	重量部							20	20
(D) ガラスビーズ	重量部	30	30						
(D) マイカ	重量部			30	30			10	10
(D) ガラスバルーン	重量部					30	30		
Izod衝撃強度	kgfcm/cm	9.8	2.0	9.5	1.6	9.5	1.9	16.6	4.6
表面状態試験	—	○	×	○	×	○	×	○	×

[Translation done.]

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CLAIMS

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[Claim(s)]

[Claim 1] (A) Thermoplastic polyacetal resin 100 It is (B) to the weight section. Acid-anhydride denaturation polyolefine system resin 1-100 The weight section and (C) The esterification reaction catalyst 0.01 - 5 weight sections, and (D) One or more sorts of bulking agents 0-150 chosen from a fibrous bulking agent and powder-like bulking agent, a tabular bulking agent, and a hollow-like bulking agent Polyacetal resin constituent which blends the weight section and changes.

[Claim 2] (B) Acid-anhydride denaturation polyolefine system resin is polyolefine system resin 100. Polyacetal resin constituent according to claim 1 which denaturalizes the weight section in an acid anhydride 0.1 - 30 weight sections.

[Claim 3] (C) The polyacetal resin constituent according to claim 1 or 2 which is one sort chosen from the group which an esterification reaction catalyst becomes from pyridines, its derivative or those salts and tertiary amine, or its derivative, or two sorts or more.

[Claim 4] (C) The polyacetal resin constituent according to claim 1 or 2 which is one sort chosen from the group which an esterification reaction catalyst becomes from quaternary ammonium salt or its derivative, an aliphatic series organic acid, aromatic series organic acids or those salts, an organometallic compound, a zinc chloride, magnesium titanate, and magnesium zirconate, or two sorts or more.

[Claim 5] (A) For polyacetal resin, the content of the hydroxyl group in resin is 30 mmol/kg. Polyacetal resin constituent of claim 1-4 which is the above thing given in any 1 term.

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[Translation done.]